Microstructural analysis of geopolymer pastes contaminated with crude oil: Impact on efflorescence, physical properties, and mechanical performance

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ABSTRACT

Geopolymer concrete represents a more sustainable alternative to traditional cement in particular production processes, reducing carbon emissions, recycling industrial by-products, and promoting energy efficiency. However, a notable challenge is the potential for efflorescence, which can impair material performance due to microstructural changes from alkali leaching and carbonation. Various additives have been explored to mitigate efflorescence. Using oil-contaminated sand in geopolymer and concrete cement production is emerging as a cost-effective rehabilitation strategy that also addresses environmental concerns. This study investigates the impact of crude oil contamination on geopolymer pastes, focusing on efflorescence susceptibility. Under normal ambient conditions, the geopolymer pastes did not show signs of efflorescence. However, exposure to water led to the formation of efflorescence-type products. Notably, crude oil contamination significantly influenced this outcome. Crude oil significantly decreased the occurrence of efflorescence, mainly when contamination levels ranged from 2 % to 10 %. This reduction is attributed to the oil's effects on the paste's pore structure, fluid migration dynamics, and viscosity, which affected water adhesion and movement through the paste. Additionally, crude oil contamination altered the chemical properties of the geopolymer pastes, causing fluctuations in the pH of leaching solutions, which affected chemical equilibrium and electrical conductivity. Compressive strength improved with 1 % crude oil contamination but decreased with higher contamination levels. Geopolymer paste with 1 % crude oil content generally increases material density and reduces porosity, enhancing mechanical properties and reducing efflorescence. However, excessive contamination undermined these benefits and negatively impacted material performance. Therefore, moderate oil contamination can be effectively used as an additive to reduce efflorescence and enhance geopolymer concrete's physical and mechanical properties. The findings provide valuable insights into the potential applications and limitations of using fly ash-based geopolymers in environments prone to oil contamination.

1. Introduction

Crude oil is one of the most essential energy sources and plays a significant role in improving the economy of any country. However, the side effect of oil contamination resulting from accidental oil spills is one of the essential impacts on the environment [1]. Despite several remedial strategies to mitigate the ecological consequences of crude oil pollution, they are not considered economically feasible. [2,3]. The

mechanical characteristics of contaminated sand and crude oil pollution must be considered while choosing the most cost-effective and efficient restoration strategy. Eagle, et al. [4], presented the capital cost of the cheapest remediation methodwitch is soil washing. The remediation process took 23 months and involved attrition, screening, and wet classification at the soil-washing plant. On the other hand, the potential benefits of using contaminated sand in construction have been investigated in research studies. Ajagbe, et al. [5] conducted a study to examine

how crude oil affected the compressive strength of concrete. They concluded that 2.5–25 % of the crude oil contamination resulted in the loss of 18–90 % of the compressive strength. In a recent study by Abousnina, et al. [3], the feasibility of using oil-contaminated sand in the production of geopolymer cement mortar was thoroughly investigated. The research concluded that geopolymer mortar can effectively incorporate oil-contaminated sand, which repurposes waste material and significantly reduces the environmental impact of sand disposal and cement production. This approach highlights a sustainable method for managing contaminated sand and contributes to environmental conservation in the construction industry. However, geopolymers have gained significant recognition for their impressive mechanical performance and ability to minimise environmental effects by emitting low carbon dioxide (CO_2) [6–9].

The extensive adoption of these materials is still constrained by the scarcity of abundant, uniform, affordable, and viable raw materials in certain areas, the absence of global benchmarks for their manufacturing and assessment, and an incomplete comprehension of their long-lasting characteristics of alkali-aggregate reaction, carbonation, acid corrosion, efflorescence, and the fundamental physicochemical interactions that govern these behaviors [7]. Consequently, increased durability is a significant factor that motivates the acceptance and use of this technology. Considerable attention has been given to studying the durability of geopolymers in various adverse conditions, such as marine environments, exposure to sulphates and chlorides, and carbonation [10–12]. Nevertheless, the precise regulation of the mixture proportions that determine the vulnerability of geopolymers to alkali leaching and consequent efflorescence when in contact with water remains incompletely comprehended. Specifically, a successful approach for removing efflorescence is lacking in the available research [13]. Efflorescence occurs when water comes into touch with the pore structure and causes the leaching of the free alkalis. The alkalis leached react with carbonic acid, produced by dissolving ambient (CO_2). This reaction results in the formation of alkali salts, which subsequently crystallise on the surface of the geopolymer [12,14].

Efflorescence has also been observed in Portland cement [15]. However, geopolymer cements are particularly susceptible to this phenomenon due to their higher alkali concentrations, which may not be fully chemically bonded within the gel structure. Research indicates that the pore structure significantly influences the movement of free alkalis in the pore solution—resulting from excess, weakly bonded, or unreacted alkalis—along with their extraction during leaching. A denser and more tortuous pore network tends to inhibit the movement of alkali cations. The production of efflorescence is further influenced by physical and chemical properties that depend on the synthesis conditions and proportions of the geopolymer mix [16,17]. To mitigate efflorescence, the alkali content of the cement must be reduced, and the solubility and absorption rate of CO_2 from the environment must be minimized [13]. Literature indicates that geopolymer mixes can experience rapid efflorescence when dried surfaces are exposed to water and humid conditions. Various factors, such as the type of alkali metal, raw materials used, and specific reaction conditions, have been shown to affect the degree of efflorescence in geopolymer concrete [18–20]. However, challenges such as the lack of global benchmarks for manufacturing and characterization, inconsistent availability of suitable raw materials in certain regions, and a limited understanding of durability properties—including resistance to acid attack, carbonation, and alkali-aggregate reaction—pose barriers to broader adoption of these materials [21]. Upon contact with water, unbound alkalis within the pore structure can lead to leaching and subsequent efflorescence. This process involves alkalis reacting with carbonic acid, formed when ambient carbon dioxide dissolves, resulting in alkali salts that crystallize on the geopolymer's surface [22]. This study aims to evaluate efflorescence formation in various geopolymers from fly ash with differing percentages of crude oil contamination. By intentionally varying the degree of contamination, we will correlate efflorescence formation with

the gel's physical, chemical, and microstructural properties.

2. Experimental program

2.1. Materials and sample preparation

Type F (low calcium) fly ash was used in this study. This was sourced from Pozzolanic Millmerran in Queensland, Australia. The chemical composition of the fly ashes is given in Table 1. The density of fly ash is 1100 kg/m^3. Extensive research has been conducted on the preparation of Millmerran fly ash over several years, with the composition design of these materials detailed in previous studies referenced in the literature [3,23,24].

Alkali activators were analytical grade NaOH (~99 %) dissolved in water and a sodium silicate solution with 29.4 wt% SiO_2, 14.7 wt% Na_2O, and 52.7 wt% H_2O, supplied by PQ Australia. The composition of the alkali activator was carefully adjusted by blending sodium hydroxide (NaOH) and sodium silicate solutions to achieve the desired molar ratios necessary for optimal geopolymerisation. This sodium silicate and sodium hydroxide combination is the alkaline liquid required to activate the fly ash or other precursor materials. The specific gravity of the alkaline solution is influenced by its concentration, measured in terms of molarity (M). Molarity refers to the number of moles of solute per liter of solution, and it plays a crucial role in determining the effectiveness of the activator in promoting the chemical reactions that form the geopolymer matrix. The alkaline solutions were prepared to ensure proper activation by mixing the sodium hydroxide and sodium silicate at least one day before mortar mixing. This advanced preparation allows the solutions to integrate fully, ensuring a uniform mixture that can effectively activate the precursor materials during the geopolymer formation. By allowing the solutions to sit, any exothermic reactions that may occur when combining the two solutions can stabilize, leading to a more consistent and reliable activator for the geopolymer paste. This careful preparation is essential for achieving the desired properties and performance of the final geopolymer product. The concentration for making geopolymer concrete typically ranges from 8 M to 16 M. A study by Hardjito and Rangan [25] indicated that a 12 M NaOH solution contains 361 g of NaOH per kg of solution. The specific gravity of this solution is calculated to be 1.10, based on the actual weight of a 1 L solution being 1329.6 g. Therefore, a 12 M solution was prepared for this study. The samples were prepared by mixing fly ash (the binder) with nine different percentages of light crude oil: 0.5 %, 1 %, 2 %, 4 %, 6 %, 8 %, and 10 % by weight. An uncontaminated sample (0 %) was also prepared as a control. The composition of the geopolymer mortar adhered to the AS 2350.12 2006 standards. Mineral Fork W2.5 motorcycle oil was used to contaminate the aggregate, as its density and viscosity closely resemble those of light crude oil [3]. This study combined fly ash (FA) with alkaline liquid (AL) before introducing oil. This methodology is based on prior research that explored the effects of different mixing methods [26]. The pastes were then mechanically mixed for 5 minutes and cured at 60°C for 24 hours. Following this, they were stored in sealed plastic containers at approximately 25°C with a relative humidity of at least 90 % for 28 days before exposure.

2.2. Tests conducted

Efflorescence formation was assessed after 28 days of curing, subjected to crude oil contamination levels ranging from 0 % to 10 %. Using cylindrical samples (30 mm diameter and 60 mm height), the paste was assessed under two conditions: partially immersed in water and stored at room temperature. To promote efflorescence, samples were immersed to a depth of 4–5 mm in water at 20 $\pm$ 5 °C and 50 $\pm$ 15 % relative humidity. Evaluations were conducted at 7, 28, and 56 days to monitor changes and assess the effects of contamination, with additional water added daily to maintain the initial water level.

Determination of leached sodium: The pH of this solution was

Table 1
Chemical composition of fly ash (%).

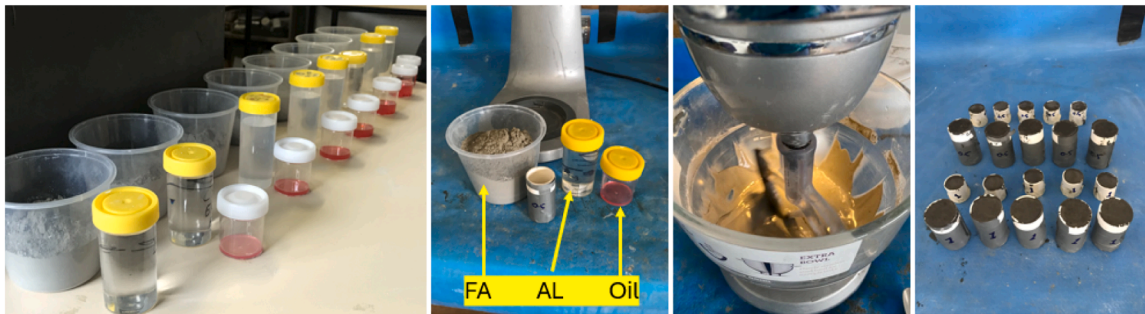| Element	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3
Percentage (%)	51.8	24.4	9.62	4.37	1.5	0.34	1.41	0.26

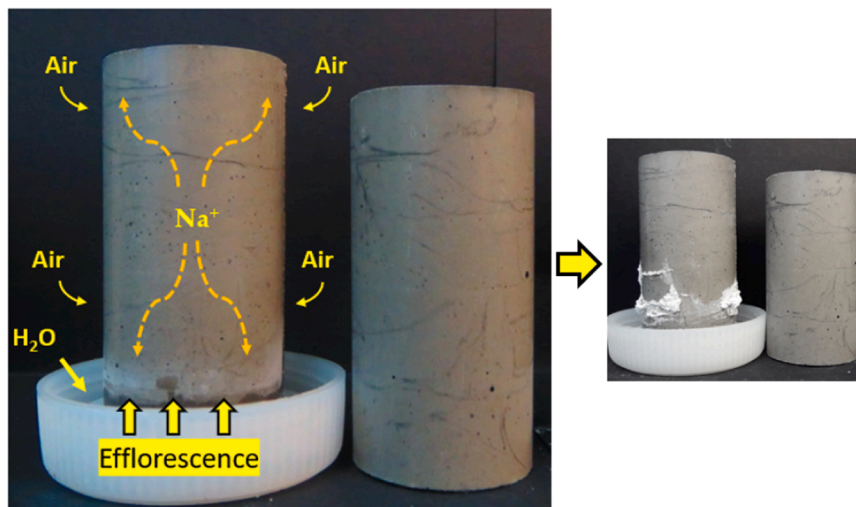**Fig. 1.** Sample preparation (geopolymer paste with different crude oil content 0 %-10 %.

Fig. 2. Efflorescence formation experiment.

measured at 28 days. a cylindrical sample was immersed in 500 ml of distilled water, and the pH was measured at 28 days. Additionally, pH measurements were taken every 10 minutes for the first hour, hourly for the first 6 hours, and then daily for 30 days, using a Eutech PC 2700 pH and conductivity analyser.

Capillary water absorption was quantified using the capillary absorption method, where the dried cylindrical samples $\Phi25\times$ 25 mm were partially immersed in 5 mm of water. The amount of absorbed water was measured every 10 min in the first hour and then every hour to achieve constant mass. Two samples of each crude oil contamination were merged in 5 ml of H_2O, and the water absorption was recorded gradually up to 56 days.

Isothermal calorimetry The geopolymerization reaction in fresh pastes was monitored using a TAM Air microcalorimeter with a sensitivity of ±20 mW. The calorimeter was set at 60 °C, and the precursor and alkali activator solution were pre-heated for 40 minutes before mixing. Internal mixing was applied for 2 minutes to prevent heat loss during the geopolymerisation reactions. The AL/b ratio was adjusted to 0.50 during calorimetry tests for a complete and homogeneous mixture.

Porosity In this study, a helium porosimeter was employed to measure the porosity of the samples. This instrument accurately determines the pore volume and size distribution by analysing the adsorption of helium gas, which can penetrate even the smallest pores

due to its low atomic size. Using the helium porosimeter, we obtained detailed insights into the porosity characteristics of the samples, including total porosity and the distribution of pore sizes. This information is crucial for understanding the investigated materials' structural properties and performance implications.

Density Alfa Mirage Electronic Densimeter Conform to Pycnometer and Hydrostatic Method Model:n SD-200L ASTM D297–93–16 was used to determine the density of all specimens with different crude oil contaminations.

2.3. Microstructure analysis SEM

The microstructure of all the samples was observed using a scanning electron microscope (SEM) (JEOL JCM-6000, Tokyo, Japan), to investigate the effect of light crude oil. All the specimens were cut, polished, and coated with a thin layer of gold-palladium using a vapor-deposit process before using SEM [27].

X-ray diffraction (XRD) analysis was conducted on geopolymer pastes with varying crude oil content (0–10 %) using a Bruker D8 Advance diffractometer, operating at 40 mA and 35 kV. The setup included an axial convolution source length of 12 mm, primary and secondary Soller slits, a step size of 0.013°, a goniometer range of 5–120°, and a counting time of 0.7 seconds.

The compressive strength of the hardened was determined at 28 days. Each cylindrical sample was cast in a PVC pipe, with nominal dimensions of 25 mm in diameter and 25 mm high, stored at room temperature for 24 h, then extracted from the mould and stored again at room temperature for another 28 days. Before testing, the specimens were surface ground using a grinding machine to ensure smooth cross-sectional surfaces. Once complete, each cylinder exhibited a highly polished surface, contributing to improved load distribution, shape, and dimension accuracy. The compressive strengths of the samples were determined following the test procedure prescribed in ASTM C579 [28], by the universal testing machine with a capacity of 100 kN. The loading speed was 0.5 mm/min.

3. Results and discussion

3.1. Effect of crude oil on the efflorescence formation

Fig. 3 displays the visual assessment of geopolymer paste exposed to varying degrees of crude oil contamination (ranging from 0 % to 10 %). The paste was tested under two conditions: partially emerged in water and at room temperature. The evaluations were conducted at different intervals (7, 28, and 56 days). The geopolymer pastes did not exhibit efflorescence during the 56 days under ambient conditions (AC). However, when exposed to water, samples formed efflorescence-type products on their surfaces, resulting in surface efflorescence [20]. After 28 days, the sample with no oil contamination exhibited significant efflorescence, while samples with 0.5 % and 1 % oil contamination were the next to show noticeable efflorescence, but this occurred only after 56 days. As the concentration of crude oil increased, the amount of efflorescence decreased. However, only a minor efflorescence was observed in all samples with 2 % to 10 % crude oil contamination, even after 56 days, as shown in Fig. 3. This suggests that higher crude oil concentrations effectively inhibit rapid efflorescence. Contrary to traditional views, a porous structure typically facilitates the movement of water and alkali cations through additional pathways, which can encourage carbonation [3]. However, higher concentrations of crude oil can delay efflorescence once the pores are filled with oil. Crude oil's interaction with microscopic pore structures affects fluid migration. Basheer [29] found that crude oil occupies pores and decreases water flow due to its hydrophobic nature, which blocks capillary action. Furthermore, crude oil alters water adhesion properties on mineral surfaces. Oil reduces water's surface tension and adhesion, restricting its movement through material pores and limiting the diffusion of salts essential for efflorescence formation [30,31]. Moreover, the viscosity of crude oil compared to water impacts water migration rates. High-viscosity crude oil significantly slows fluid movement within pores, thereby delaying efflorescence formation by reducing the rate at which dissolved ions reach the material's surface [32].

3.2. Impact of leachability on oxide composition

Fig. 4 shows the pH change of the leaching solutions. At the static leaching conditions, the pH value increases rapidly from 9.0 to 9.8 at three hours to 11.0–11.3 at 24 hours, followed by a slower increase to 12.0–12.3 until 168 h. Afterward, the pH values for each of the mixtures remain almost unchanged. The pH value measurement results agree with the sodium leaching trend. Crude oil is a complex mixture of hydrocarbons and other organic compounds that can have varying effects on construction materials. Research has shown that organic contaminants can alter the chemical equilibrium of concrete systems, resulting in pH fluctuations [33]. When geopolymer concrete comes into contact with crude oil, the oil can penetrate the porous structure, interacting with the alkaline components and potentially reducing the pH level [34].

3.3. Electrical conductivity of leaching solutions

Electrical conductivity plays a significant role in the performance of geopolymers, influencing their material properties, durability, thermal characteristics, and environmental applications [35]. It indicates ionic mobility, with higher conductivity often correlating to a greater presence of soluble ions, which can affect mechanical strength and durability in varying moisture conditions [36]. Conductivity measurements can also assess a geopolymer's resistance to environmental factors, such as acids and alkalis, with lower conductivity linked to improved chemical resistance [37]. Additionally, higher ionic conductivity can enhance thermal stability and is crucial in remediation efforts to immobilize contaminants [38]. On-site, understanding and monitoring electrical conductivity can aid in predicting material behavior in diverse environmental conditions, provide real-time quality control during mixing and curing, and support long-term performance assessments, allowing engineers to make informed decisions about material selection [39]. Fig. 5 presents the electrical conductivity of the leaching solution of geopolymer pastes with different crude oil content (0–10 %). The conductivity increases sharply with the leaching time during the 1440 minutes. However, the electrical conductivity of the leaching solution slightly decreased after 24 hours up to 7 days, and it was almost constant between 7 and 28 days. Compared with the pH testing results, the electrical conductivity variation trend is consistent with the pH variation during all the leaching stages (up to 28 days). The electrical conductivity of the control sample fits the same trend as pH. Furthermore, the electrical conductivity of the leaching solution of geopolymer pastes decreased as the amount of crude oil increased. For instance, the electrical conductivity of the leaching solution of geopolymer pastes decreased by 6.7 % and 4.06 % when 10 % of crude oil content was used compared to the control sample at 7 and 28 days. It can be said that the crude oil's presence within a geopolymer structure can alter the conductive pathways by creating barriers for ion movement or introducing additional ions into the leaching solution. The type and composition of crude oil can have differential effects on the conductivity profile of leaching solutions [40]. For instance, lighter oils with more volatile compounds may lead to different conductivity outcomes than heavier oils with larger molecular structures [41]. Crude oil, a complex mixture comprising various hydrocarbons and other compounds, can infiltrate geopolymer concrete through these porous networks, establishing physical and chemical barriers to ion movement [42]. Physical barriers result from oil's occlusion of pore spaces, which obstructs the pathways necessary for ion diffusion. On a chemical level, components within the crude oil can interact with the geopolymer matrix, altering its original chemistry and potentially hindering the ion exchange required for geopolymerisation and maintaining structural integrity. In general, crude oil content reduces the leaching rate in the period studied.

3.4. Capillary water absorption

The amount of water absorbed by capillarity for each sample is shown in Fig. 6. In systems with samples that were not contaminated, the mass steadily grew up to 48 hours. Then, it stayed unaltered beyond this point, which indicates that the saturation point had been reached during this period. In contrast to the control sample, the samples containing 0.5 %, 1 %, and 2 % concentrations reached the saturation stage at 96 hours, slightly delayed compared to the control sample. A considerable decrease in water absorption was seen in the samples as the percentage of crude oil grew from 4 % to 10 %. It was found that the geopolymer samples with 8 % and 10 % crude oil contamination had the lowest values compared to the control sample, which were approximately 7 g/cm^2. The water permeability value of the samples with 6 % crude oil contamination was about 8.5 g/cm^2, while the samples containing 4 % had a bit higher value of approximately 9.2 g/cm^2. At the beginning of the experiments, the absorption rate was much slower for every sample with crude oil contamination. As the amount of crude oil
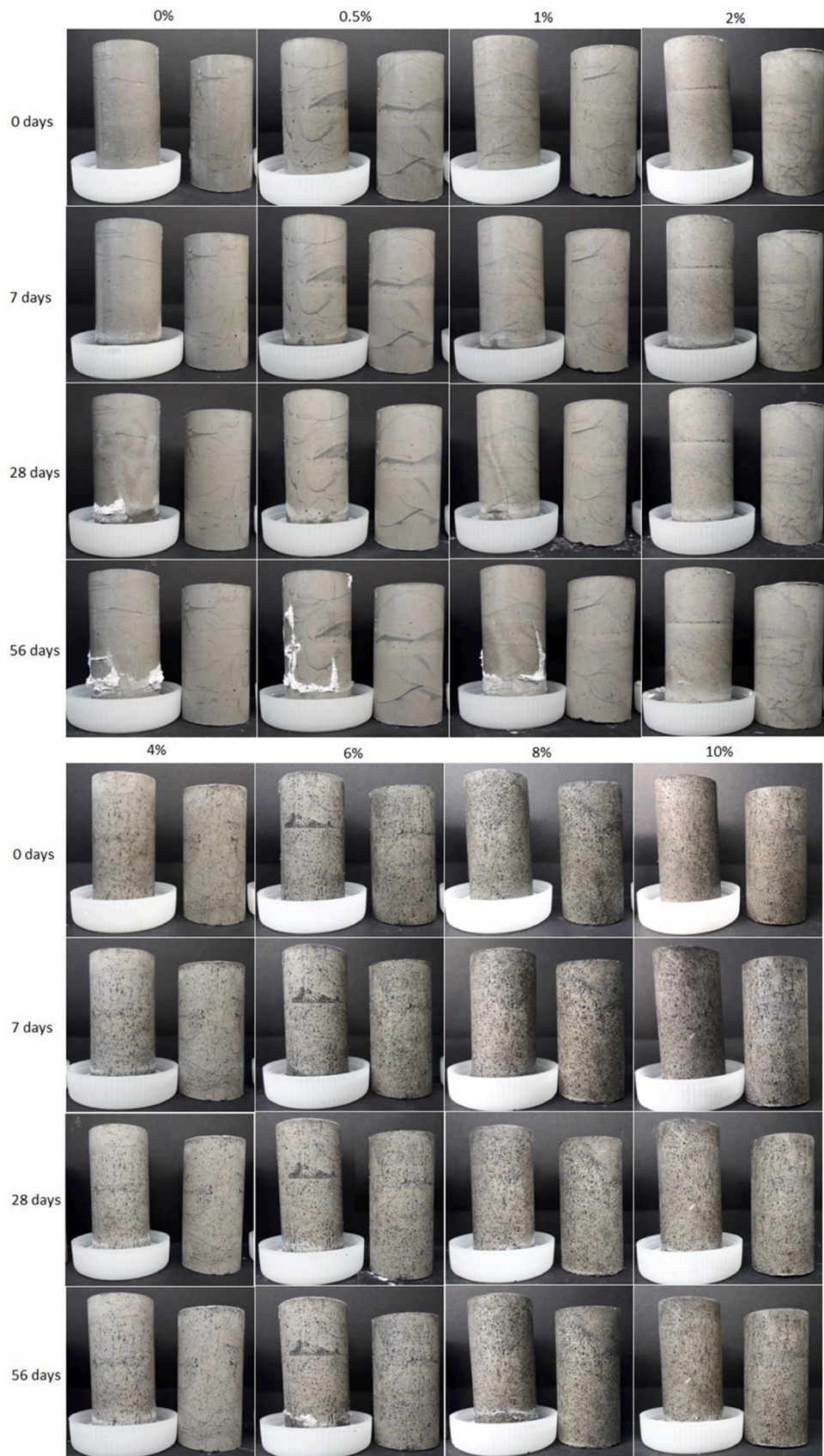
Fig. 3. Visual evidence of efflorescence in geopolymer samples with different crude oil content.
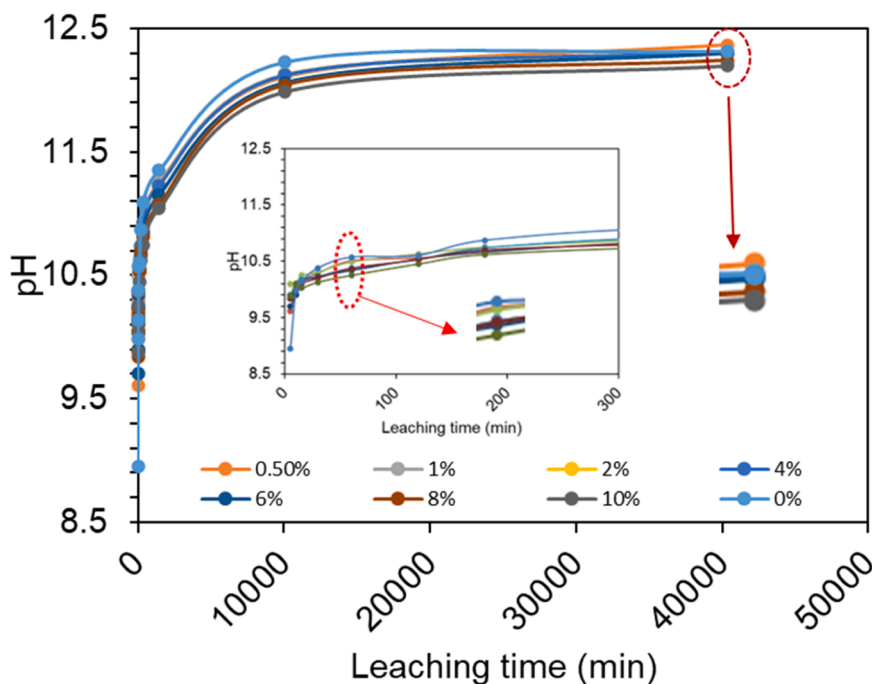
Fig. 4. pH of the leaching solutions for geopolymer paste with different crude oil content.
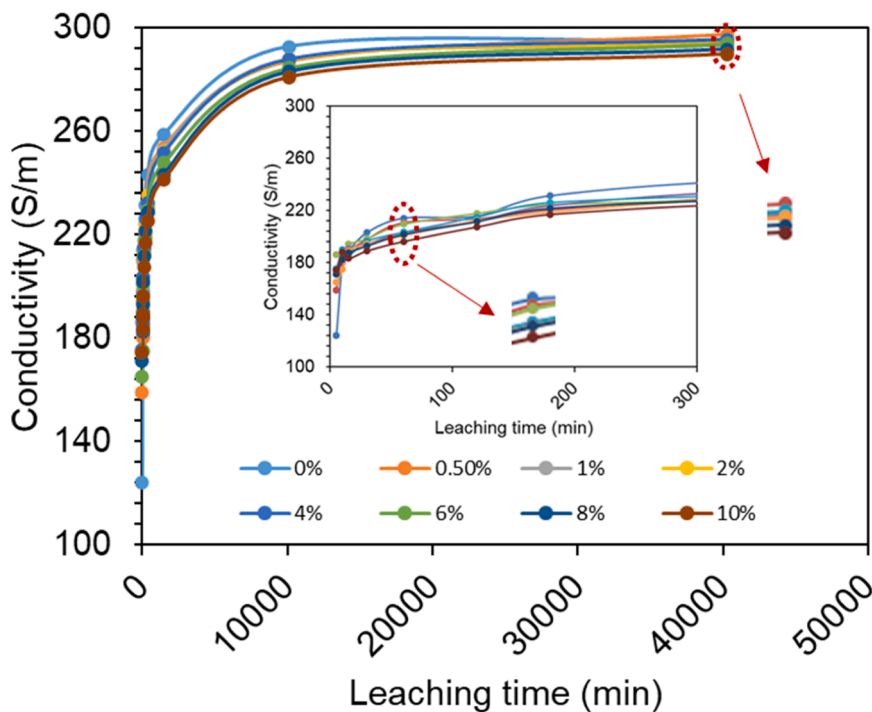

Fig. 5. Electrical conductivity of the leaching solution of geopolymer paste with different crude oil content (0 %-10 %).

steadily rises, the amount of absorbed water gradually decreases. However, after 48 hours, samples 0.5 %, 1 %, and 2 % revealed values comparable to those uncontaminated samples. There is a clear correlation between the trends of water absorption and the measures of pH, the degree of alkali leaching, and the visual observation of efflorescence. As a result, the degree of water permeability is also a significant factor in determining the level of alkali leaching and, therefore, efflorescence in these systems. Crude oil significantly impedes the permeability of geopolymers, affecting their ability to transport substances and facilitate

water flow. Several studies have demonstrated this effect, where crude oil acts as a barrier within the porous structure of geopolymers, thereby restricting water movement through the material. For instance, Adjei, et al. [43], demonstrated that crude oil alters the pore structure of geopolymers, reducing their effectiveness in water transport. This hindrance is attributed to the hydrophobic nature of crude oil, which interacts with the hydrophilic matrix of geopolymers, leading to decreased water permeability [3]. These findings underscore the importance of considering environmental contaminants such as crude oil when
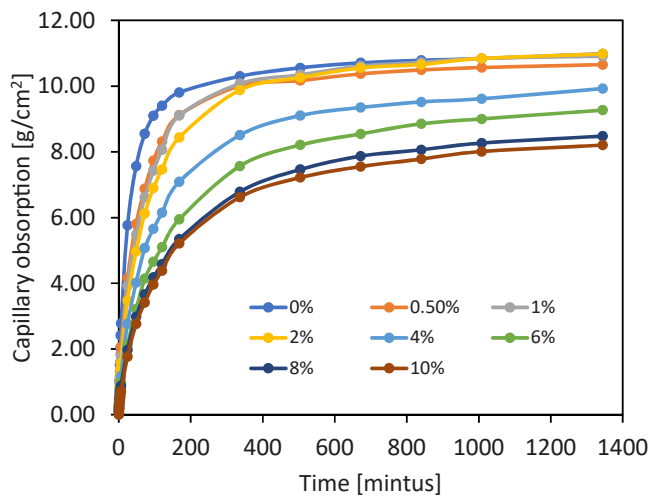
Fig. 6. Capillarity absorption of geopolymers paste with different crude oil content.

assessing the performance and durability of geopolymers in practical applications.

3.5. Heat release of geopolarisation process

Fig. 7(a-b) shows geopolymer pastes' heat flow and cumulative heat curves with different crude oil content determined by the isothermal calorimetry at 60∘C. A noticeable exothermic peak is present in all geopolymer pastes with different crude oil contamination, primarily associated with the interaction and dissolution of the solid precursors and the alkaline solution. During the initial stage of the polarisation process, the combination of rapid chemical reactions, heat release from polymerisation, and structural transformations leads to higher exothermic peak values observed in the thermal analysis of the samples. The presence of one exothermic peak in geopolymerisation is due to its simpler chemical composition and more uniform reaction kinetics compared to the multi-phase hydration process of cement, which results in multiple exothermic peaks [44]. The data shows that the total heat released increased by adding crude oil content to the geopolymer paste. Specifically, the paste samples contaminated with 1 % crude oil exhibited a notable increase in total heat released, rising by 3 % compared to the control sample. Conversely, samples with 0.5 % crude oil contamination showed a more modest increase of 0.5 % in total heat released than the control. However, beyond 2 % of crude oil contamination, heat release during paste formation decreased with increased crude oil content. This suggests that higher levels of crude oil contamination may negatively impact the heat release during geopolymerization beyond a certain threshold. The cumulative heat release during geopolorisation is an important parameter that influences the reaction's kinetics and the material's final properties. It measures the energy released over time as the geopolymer cures. Researchers can infer the geopolymer's reaction rate and mechanical strength development by monitoring the heat flow.

The observed increase in both cumulative heat and heat flow at lower levels of crude oil contamination (0.5 % and 1 %) can be attributed to a reduction in the total surface area of the fly ash (FA) binder compared to the alkaline solution (AL). As illustrated in Fig. 8, at oil content of 0.5 % and 1 %, crude oil partially covered the fly ash particles, leading to a decrease in the accessible surface area. This partial coverage likely facilitated a more efficient alumina and fly ash reaction, enhancing reactivity and heat release. The reduced surface area of the binder and improved interaction between AL and FA led to greater density and reduced porosity in the geopolymer paste. Consequently, samples with this oil content were denser than the control samples.

These improvements are reflected in the enhanced mechanical properties and diminished efflorescence observed in the contaminated samples. Consequently, the optimal reaction conditions and modified physical properties at these content levels suggest that the presence of crude oil, up to a certain threshold, may positively influence the geopolymerization process by modifying binder characteristics and enhancing performance.

Conversely, increased crude oil content from 2 % to 10 % resulted in a notable decrease in cumulative heat and heat flow during the geopolymerization process. For example, the geopolymer paste with 1 % crude oil contamination exhibited a maximum exothermic peak of 8.12 mW/g, while the paste with 0.5 % crude oil contamination showed a slightly lower peak of 8.0 mW/g. In comparison, the control samples, which had no crude oil contamination, displayed a peak of 7.9 mW/g. However, as the crude oil content progressively increased from 2–10 %, a gradual decline in heat flow was observed. Specifically, the heat flow decreased to 7.8 mW/g at 2 % crude oil, 7.7 mW/g at 4 %, 7.6 mW/g at 6 %, 7.5 mW/g at 8 %, and 7.2 mW/g at 10 % crude oil contamination. This decrease in heat flow with higher crude oil content indicates a diminishing reactivity and heat release efficiency, which may be attributed to the excessive crude oil interfering with the chemical reaction between the fly ash and alumina. The results suggest that while low levels of crude oil can enhance the geopolymerisation process by optimising binder properties, higher concentrations may inhibit the reaction, leading to reduced thermal activity and potentially impacting the overall performance of the geopolymer material. Crude oil consists of a wide range of organic compounds, including aliphatic, aromatic, and heteroatomic hydrocarbons, whose presence during geopolymerisation can introduce multiple variables. According to Ahmaruzzaman [45], the chemical interactions between crude oil components and the alkaline activator can alter the pH of the solution, which is a controlling factor in the dissolution rate of aluminosilicate materials. Hence, the presence of crude oil can indirectly influence the initial stages of geopolymerisation and the corresponding heat release. Furthermore, the research of Ismail, et al. [46] supports these findings, where it indicated that the interfacial tension between crude oil and the alkaline solution affects the oil dispersion in the geopolymer matrix. This dispersion can create micro-encapsulations of oil, which can act as physical barriers, modifying the heat flow pathway during curing [47,48]. A study by Montazeri and Eckelman [48] involved the preparation of geopolymer samples with varying concentrations of crude oil. The researchers found that low levels of crude oil contamination did not significantly impact the cumulative heat release. However, there was a noticeable reduction in heat release at higher concentrations compared to uncontaminated samples, suggesting that crude oil components interfered with the polymerisation process [49]. Further evidence comes from a paper by Najafi Kani, et al. [50], which measured the cumulative heat release using isothermal calorimetry at different stages of the geopolymer curing process. Their results revealed that initial heat flow rates were lower in samples with crude oil, confirming that the oil affects the early stages of reaction kinetics. The presence of crude oil has been shown to modulate this heat release, affecting the performance and suitability of geopolymers for specific applications. While low concentrations of crude oil may exert negligible influence, higher concentrations have been empirically proven to reduce the cumulative heat release during geopolymer curing. The presence of crude oil at high concentrations (greater than 2 %) has modulated heat release during the geopolymerization process, thereby impacting the performance of geopolymers. While low concentrations of crude oil can enhance heat flow, higher concentrations have been empirically demonstrated to decrease cumulative heat release during the curing of geopolymers. This decrease in heat release can negatively affect the overall performance and effectiveness of the geopolymer material.
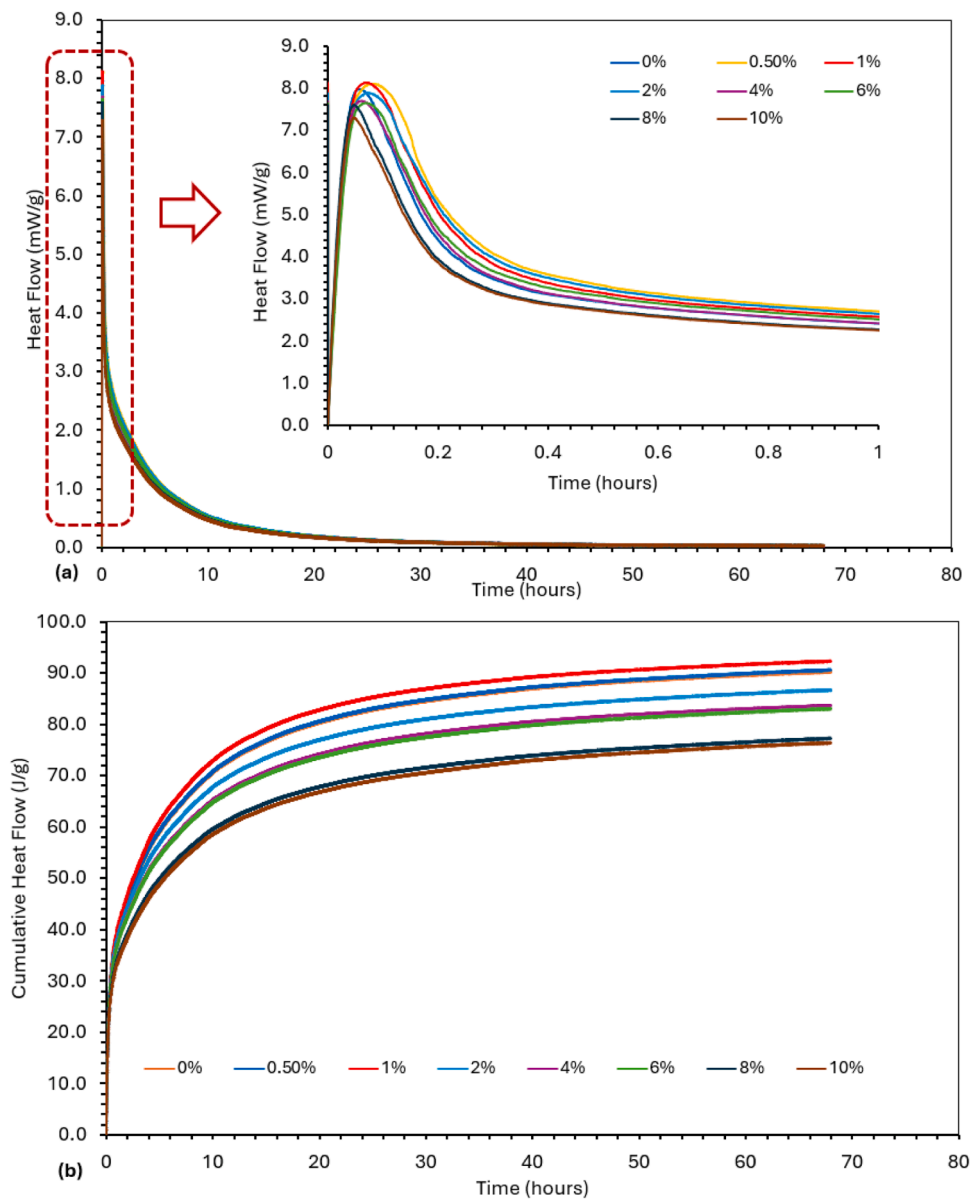
Fig. 7. Geopolyrisation process of geopolymer with different crude oil content: (a) Heat flow, (b) Cumulative heat.

3.6. Compressive strengths

The effects of different crude oil content on the compressive strength of the geopolymer paste are shown in Fig. 9. At 0.5 % crude oil content, the compressive strength of the geopolymer paste was 0.5 % higher than control samples (uncontaminated samples). While at 1 % oil content, the strength gain of geopolymer paste was 10 % higher than the control samples. On the other hand, there was a more significant decrease in the compressive strength of geopolymer paste when the oil content was 2 % or higher. The minimum compressive strength was obtained with 10 % crude oil content (13.8 MPa). The increased compressive strength at 0.5 % and 1 % crude oil contents indicate that these concentrations may optimize the geopolymerisation process. This enhancement in strength is likely due to a reduction in the effective surface area of fly ash (FA) particles exposed to the alkaline activator, as illustrated in Fig. 8 above. The oil partially coats the FA particles at these crude oil levels, modifying their interaction with the alkaline solution. This partial coating creates a more controlled reaction environment, facilitating more effective geopolymerisation. Consequently, this controlled environment enhances mechanical performance, as evidenced by the increased

density and reduced porosity of the geopolymer samples compared to the control sample. The observed higher compressive strength reflects these optimized conditions' advantages. These results are further supported by additional analyses, including porosity measurements and geopolymerisation processes such as heat flow and cumulative heat, which are discussed in detail in Section 3.5. Overall, the presence of crude oil likely contributes to improved strength and durability of the final geopolymer product. Researchers have reported that the activity of the pozzolanic reaction may be enhanced by reducing the total surface area of fly ash particles [51,52]. This is probably caused by a reduced need for water and alkaline activators to cover the particles, leading to a denser and more cemented matrix. Furthermore, Sharma, et al. [53], found that reducing the surface area of fly ash particles may enhance geopolymers' initial and overall compressive strengths. This phenomenon is attributed to the enhanced arrangement of particles, leading to a higher matrix density. Collins and Sanjayan [54], research also revealed that reducing the surface area increased compressive strength. Collins and Sanjayan attributed this to the decreased water needed to saturate the particles, known as the water-to-binder ratio.

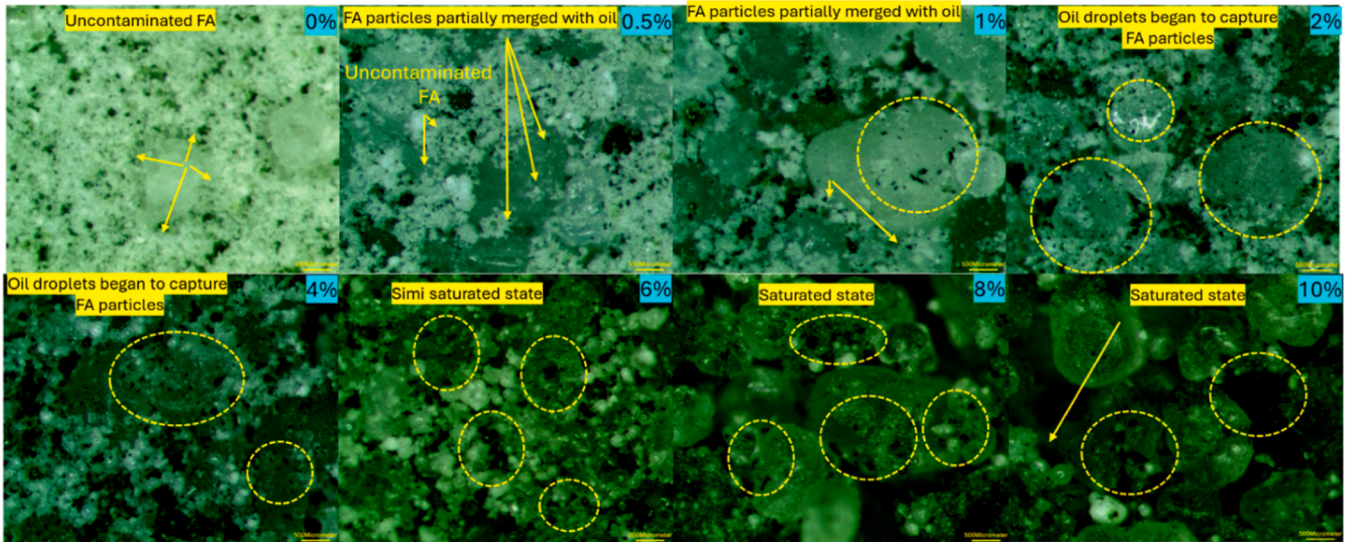Increasing the crude oil content from 2–10 % caused a significant

Fig. 8. Microscope images of a dry mix of FA and oil-contaminated sand with different %.
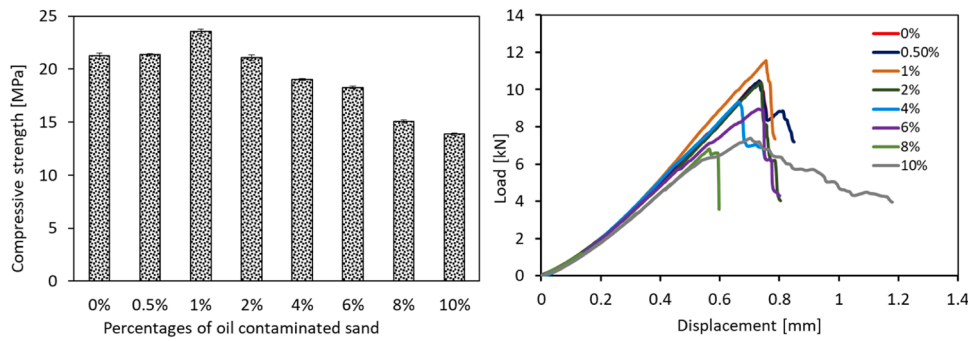

Fig. 9. Effect of crude oil content on the compressive strength of geopolymer paste (a) compressive strength (MPa) and (b) Stress vs displacement.

reduction in compressive strength due to the saturation of the fly ash and alkaline solution with crude oil. Increasing the crude oil content will increase the ratio of alkaline liquid to fly ash because the fly ash will be saturated with crude oil. Black [55], has stated that the compressive strength will decrease when the alkaline liquid-to-fly ash ratio (AL: FA) is increased above a certain level. This reduction in strength may be due to fly ash particles, with a finite surface area, not being fully saturated with alkaline liquids because the crude oil already saturates the fly ash. Hence, any more alkaline liquid is wasted within the mix at a certain point. This excess alkaline liquid then becomes extra water, diluting the mix and affecting the bond between aggregate and binder.

3.7. Density and total porosity

The density and total porosity of each sample, measured by water absorption, are shown in Fig. 10. As anticipated, a clear link between higher density and improved compressive strength was observed. Systems with 1 % geopolymer paste exhibited the highest density, which decreased progressively with increasing crude oil concentration. Conversely, porosity was lowest at 1 % crude oil content, indicating fewer voids. This behavior aligns with previous findings, suggesting that denser and stronger systems typically have lower porosity [56]. This aligns with the higher rate of water absorption and alkali leaching observed in geopolymer compared to control samples. Systems with 1 % crude oil content demonstrated higher density and lower porosity, resulting in increased strength and reduced efflorescence compared to the control samples. The inclusion of high levels of crude oil in the
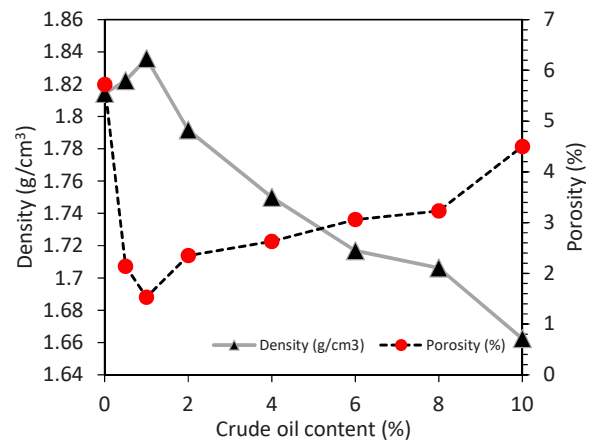

Fig. 10. Density and total porosity of geopolymers with different crude oil content.

activator not only decreased the compressive strength but also reduced visual efflorescence formation. Consequently, while compressive strength is closely associated with density and porosity, it cannot be a reliable indicator for efflorescence development. Instead, porosity and water absorption emerge as critical factors influencing the susceptibility to efflorescence [57].

The correlation between leaching potential and physical properties

indicates that the leaching process can be minimized by synthesizing a denser geopolymer material. The interplay between crude oil content and material density significantly influences mechanical properties and efflorescence. In the studied systems, density increased with the addition of crude oil up to a concentration of 1 % (1.83 g/cm^3), which is consistent with findings that higher density often correlates with improved mechanical strength [58]. The peak compressive strength was observed at this 1 % crude oil level, indicating that density enhancement benefits mechanical performance [59]. However, as the crude oil content was further increased to 10 %, a decrease in density was noted, reflecting a diminishing trend in mechanical strength. This trend aligns with previous research showing that excessive additives can disrupt the material matrix and reduce density [60]. Concurrently, the increase in crude oil content was associated with reduced efflorescence. This supports the notion that certain additives can mitigate the formation of soluble salts on the material surface [61]. Thus, while a moderate increase in crude oil content enhances mechanical properties and reduces efflorescence, higher concentrations reduce density and strength, highlighting the need for optimal additive levels in the material formulation.

3.8. XRD analysis

Fig. 11 illustrates the effect of crude oil content (0 % to 10 %) on the mineral phases formed in the geopolymer pastes. The primary crystallographic phases identified in oil-free and crude oil-contaminated pastes were Mullite, Quartz, Calcite, and Hematite. Mullite (Al4.56Si1.44O9.72), PDF No. 79–1458, was observed at Bragg angles (2θ) of 18.20, 25.52, 36.02, 42.34, and 63.33; Quartz (SiO2), PDF No. 85–0930, appeared at 22.38, 28.42, and 32.36; Calcite (CaCO$_3$), PDF No. 47–1743, was detected at 34.42; and Halite (NaCl), PDF No. 05–0628, was present at 37.32 and 52.34. These phases are consistent with those reported in the literature [62,63]. The majority phase was Mullite, followed by Quartz and Calcite, with Halite in the lowest proportion. Notably, the XRD analysis shows that increasing crude oil content did not lead to the formation of any new mineral phases. This suggests that crude oil contamination does not alter the primary crystallographic structure of the geopolymer paste.

3.9. Scanning electron microscopy

The morphologies of selected geopolymer samples after 28 days were analysed using scanning electron microscopy (SEM), as illustrated in Figure 12. The SEM images reveal that the material is homogeneous, which indicates a significant reaction between the activator and the

crude oil. The matrix appears less compact in systems with 0 % crude oil content with observable unreacted fly ash (FA) particles. In contrast, geopolymers with 0.5 % and 1 % crude oil show a robust and dense matrix with no visible unreacted fly ash (FA) particles. However, when the crude oil content exceeds 2 %, a noticeable porosity increases, leading to a less compact structure than the 0.5 % and 1 % crude oil samples. This phenomenon is consistent with reduced density and a decreased extent of reaction, attributed to a reduction in soluble silicates in the activator [22,64]. Furthermore, this increase in porosity aligns with observed reductions in mechanical performance and higher water absorption, as previously reported [65].

The more significant oil content in geopolymer paste leads to coarse pore distribution. Based on the visual observation, the pore size and the pore distribution were found to have decreased slightly for concrete with 1 % light crude oil contamination compared to uncontaminated samples (0 %). The behaviour is also consistent with the other analyses and confirms a lower porosity is observed in denser and stronger systems [22]. This observation confirms that incorporating up to 1 % crude oil significantly improved the workability of the geopolymer paste, consistent with previous findings in cement mortar studies [66]. This enhancement in workability led to a more uniform and cohesive mixture, contributing to a reduction in total porosity. Better dispersion and integration of the paste components lead to a decrease in porosity, resulting in fewer voids and a denser final product. In contrast, increasing the crude oil from 2–10 % of crude oil contamination increases the pores' sizes and distributions, as shown in Fig. 11. From the microscopic observations, the average pore size in the uncontaminated samples was 3.35 %, but only 1.53 for 1 % of crude oil contamination. This increased to 2.35, 2.63, 3.06, 3.23 %, and 4.5 % for 2 %, 4 %, 6 %, 8 %, and 10 % oil of contamination, respectively, as shown in Figs. 5 and 6. Due to the samples' saturation states and the free alkaline solution, there was an increase in porosity at high oil contamination levels. The additional crude oil boosted the porosity of the geopolymer paste, causing the pores to develop around the particles and function as reservoirs. Over-saturation led to the formation of air pockets [3]. They further indicated that fly ash carried outwards during seepage weakens the sample's surface by around 20 %. Thus, the pores appearing on the surface of the specimens could be due to the oil seeping from the geopolymer paste surface. As a result of porosity, the density of the hardened geopolymer paste decreased as the amount of crude oil increased.

4. Conclusion

The study investigates the effects of crude oil contamination on geopolymer pastes, analyzing various properties across a contamination range from 0–10 %. Key findings include:

- **Efflorescence:** The pastes initially showed no efflorescence under ambient conditions but developed efflorescence when exposed to water. Crude oil content reduced the amount of efflorescence, with minimal formation observed at 2–10 % contamination. Crude oil affects the pore structures, reducing water flow and salt diffusion and influencing water adhesion on mineral surfaces.
- **pH and electrical conductivity:** Crude oil affects the pH of leaching solutions and alters the chemical equilibrium of geopolymer systems, leading to pH fluctuations. The electrical conductivity of leaching solutions increased with time but slightly decreased after 24 hours. The type and amount of crude oil influence the conductivity profile and can establish barriers to ion movement, reducing the leaching rate.
- **Water absorption and permeability:** Crude oil contamination significantly impacts water absorption, with higher oil content reducing water permeability. Samples with 8 % and 10 % crude oil showed lower permeability than those with 4 % and 6 % crude oil. Water absorption reached saturation at 48 hours, with increased crude oil content leading to decreased absorption.
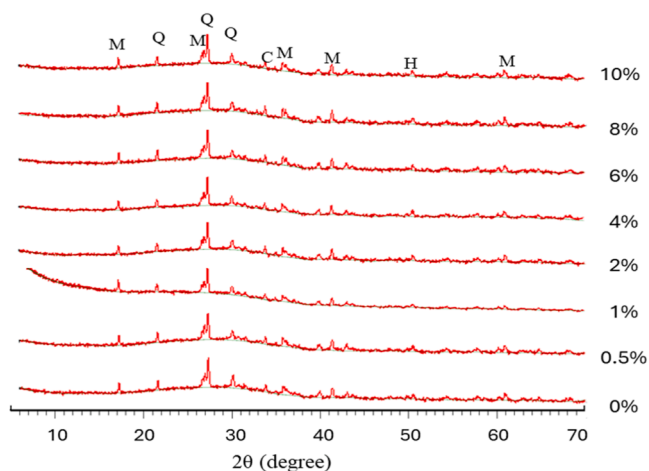

Fig. 11. XRD diffractograms of geopolymer paste with different crude oil content (0 %-10 %). Mullite (Al4.56Si1.44O9.72), Quartz (SiO2), Calcite (CaCO3) Halite (NaCl).
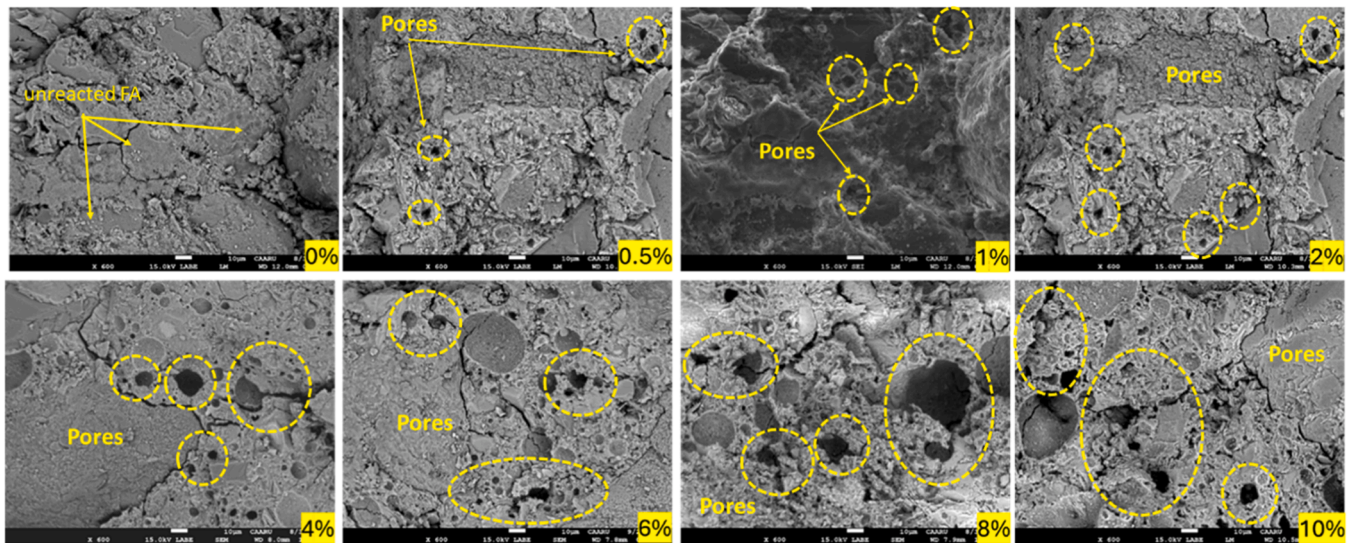
Fig. 12. Scanning electron microscopy of geopolymer pastes with different crude oil content.

- **Heat flow and geopolymerisation:** The total heat released during geopolymerisation increased with crude oil content, with a 3 % increase noted at 1 % crude oil. However, excessive crude oil may hinder the reaction, affecting heat release and overall material performance. Optimal oil content can enhance binder characteristics, but increasing the crude oil content beyond 2 % can negatively impact the reaction.
- **Compressive strength:** The compressive strength of geopolymer paste improved with 0.5 % and 1 % crude oil content but decreased at higher concentrations. Maximum strength was observed at 10 % crude oil content (13.8 MPa), likely due to fly ash particles' reduced effective surface area. Increased oil content initially improves strength, but excessive amounts can saturate the fly ash and alkaline solution, reducing strength.
- **Density and porosity:** A clear link was found between higher density and improved compressive strength. Geopolymer pastes with 1 % crude oil exhibited the highest density and lowest porosity. Higher crude oil levels decreased density and strength while reducing visual efflorescence formation. Optimal crude oil levels enhance mechanical properties and reduce efflorescence, but excessive amounts have detrimental effects.
- **Microstructural analysis:** SEM analysis after 28 days showed that pastes with 0.5 % and 1 % crude oil had a denser matrix compared to the less compact structure of those with 0 % crude oil. However, pastes with over 2 % crude oil displayed increased porosity and larger pores, resulting in higher water absorption. XRD analysis confirmed that increasing crude oil content did not form new mineral phases, indicating any alteration to the primary crystallographic structure of the geopolymer paste.

Overall, the study highlights that while moderate crude oil contamination can enhance certain properties of geopolymer pastes, excessive contamination negatively impacts density, strength, and structural integrity. Optimal crude oil levels are crucial for balancing performance and durability in geopolymer applications.

CRediT authorship contribution statement

Fahad Aljuaydi: Writing – review & editing, Resources, Investigation, Funding acquisition, Conceptualization. **Rajab Abousnina:** Writing – review & editing, Writing – original draft, Supervision, Methodology, Investigation, Formal analysis, Conceptualization. **Omar Alajarmeh:** Writing – review & editing, Visualization, Investigation.

Rafat Siddique: Writing – review & editing, Writing – original draft, Visualization, Investigation. **Abdalrahman Alajmi:** Writing – review & editing, Visualization, Investigation.

Declaration of Competing Interest

The authors declare that they have no known financial conflicts of interest or personal relationships that could be perceived as influencing the work reported in this paper.

Data availability

No data was used for the research described in the article.

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